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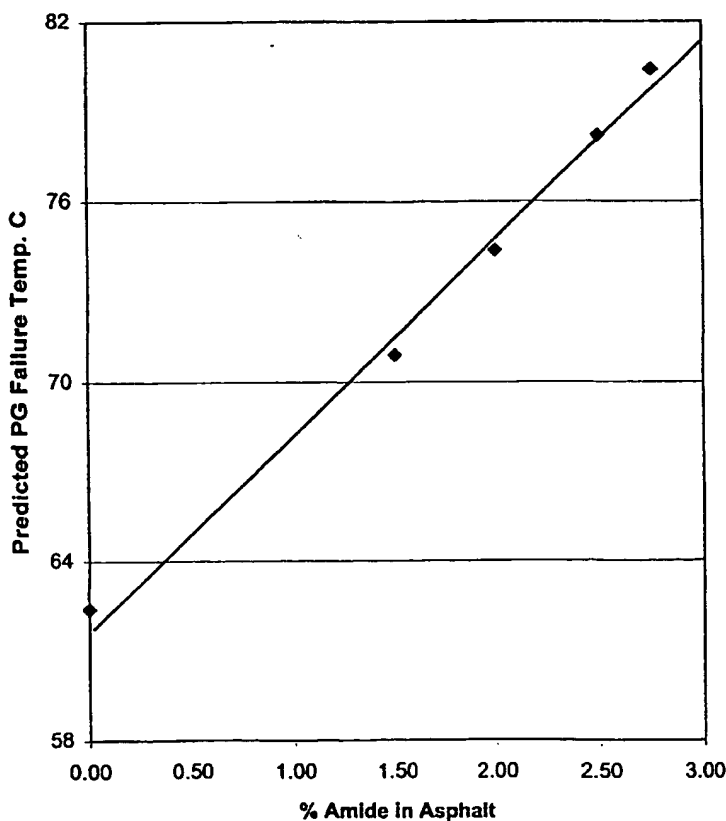
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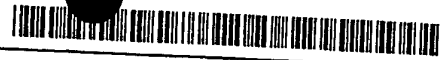
(54) Title: BITUMEN-AMIDE COMPOSITIONS USEFUL AS ROAD PAVING MATERIALS



(57) Abstract: A mixture comprising aggregate and from 2 to 8 % of a bitumen composition for road construction and repair. The bitumen composition has 75 % to 99.95 % of bitumen, 0 % to 15 % of polymer, and 0.05 % to 10 % of an amide additive that is predominately an amide having the formula:  $R_1\text{-CO-N}(\text{CH}_2)_x\text{-N-CO-R}_2$  wherein  $R_1$  and  $R_2$  are alkyl groups, each having from 12 to 52 carbon atoms, and  $x$  is an integer of from 1 to 4.

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*For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.*

## BITUMEN-AMIDE COMPOSITIONS USEFUL AS ROAD PAVING MATERIALS

The present invention relates to bitumen compositions that are useful for preparing aggregates for road construction and repair. These compositions  
5 are prepared from bitumen and amides.

### BACKGROUND OF THE INVENTION

The use of bitumen (asphalt) compositions in preparing aggregate compositions (bitumen and rock) useful as road paving materials is complicated by at least three factors, each of which imposes a serious  
10 impediment to providing an acceptable product.

First, the bitumen compositions must meet certain performance criteria or specifications in order to be considered useful for road paving. For example, to ensure acceptable performance, state and federal agencies issue specifications for the use of various bitumen applications as road pavement.

- 15 The Strategic Highway Research Program (SHRP) has developed asphalt binder specifications. The results of that program were disseminated by the Federal Highway Administration, and the American Association of State Highway and Transportation Officials has encouraged the individual states to implement those results.
- 20 The SHRP binder specifications have high temperature tests, low temperature tests, and oven aging tests. The high temperature tests determine the viscoelastic characteristics of the asphalt to control pavement rutting. The low temperature tests determine the cold flow properties of the asphalt to control low temperature cracking. The oven aging tests predict  
25 aging characteristics to estimate binder properties after extended periods of time on the road. SHRP binder grades have the form PG xxyy, where xx is the maximum pavement temperature in degrees Celsius and yy is the minimum pavement temperature in degrees Celsius. For instance, a binder grade of PG 64-28 means that the binder gives acceptable performance in  
30 the temperature range of from -28° C to 64° C.

Conventional bitumen compositions frequently cannot meet all of the requirements of a particular specification simultaneously and, if these specifications are not met, damage to the resulting road can occur, including permanent deformation, thermally induced cracking and flexural fatigue. This  
5 damage greatly reduces the effective life of paved roads.

In this regard, it has long been recognized that the properties of conventional bitumen compositions can be modified by the addition of polymers. A wide variety of polymers have been used as additives in bitumen compositions. For example, copolymers derived from styrene and conjugated dienes, such as  
10 butadiene or isoprene, are particularly useful, since these copolymers have good solubility in bitumen compositions and the resulting modified-bitumen compositions have good rheological properties.

The stability of these polymer-bitumen compositions needs to be increased by the addition of sulfur, frequently in the form of elemental sulfur. It is believed  
15 that the sulfur chemically couples the polymer and the bitumen through sulfide and/or polysulfide bonds. The addition of extraneous sulfur is required to produce the improved stability, even though bitumens naturally contain varying amounts of native sulfur.

U.S. Patent No. 3,803,066, issued April 9, 1974 to Petrossi, discloses a  
20 modified bitumen prepared by incorporating into a bitumen and co-vulcanizing with sulfur at a temperature of 125° to 145° C, the sulfur to rubber ratio being between 0.3 and 0.9.

U.S. Patent No. 4,130,516, issued December 19, 1978 to Gagle et al.,  
25 discloses an asphalt polymer composition obtained by hot-blending asphalt with 3% to 7% of elemental sulfur and 0.5% to 1.5% of a natural or synthetic rubber.

U.S. Patent No. 4,145,322, issued March 20, 1979 to Maldonado et al.,  
discloses a process for preparing a bitumen-polymer composition that contains block copolymers comprising diene and styrene groups.

U.S. Patent No. 5,019,610, issued May 29, 1991 to Sitz et al., discloses polymer-modified asphalts comprising asphalt cement and a blend of a thermoplastic rubber polymer and a fatty dialkyl amide.

5 U.S. Patent No. 5,371,121, issued December 6, 1994 to Bellomy et al., discloses asphalt compositions prepared from bitumen, a tri-block copolymer of styrene and butadiene, and sulfur.

German Application 2,331,727 discloses aggregates containing 40% to 50% methacrylate polymer, 40% to 50% bitumen, 0 to 20% stearic acid amide, 0 to 30% limestone, 0 to 40% natural asphalt, and 0 to 40% tar. In one  
10 embodiment, 50% methacrylate polymer was mixed with 50% bitumen and granulated to give an aggregate with improved adhesion and durability for rolled or cast asphalt.

The second factor that complicates the use of bitumen compositions concerns the viscosity stability of such compositions under storage conditions. In this  
15 regard, bitumen compositions are frequently stored for up to seven days or more before being used and, in some cases, the viscosity of the composition can increase so much that the bitumen composition is unusable for its intended purpose. On the other hand, a storage-stable bitumen composition would provide for only minimal viscosity increases and, accordingly, after  
20 storage it can still be employed for its intended purpose.

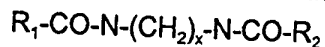
The third factor that complicates the use of bitumen compositions concerns the use of volatile solvents in such compositions. Specifically, while such solvents have been heretofore proposed as a means to fluidize bitumen-polymer compositions containing relatively small amounts of sulfur, which  
25 compositions are designed as coatings (Maldonado et al., U.S. Patent No. 4,242,246), environmental concerns restrict the use of volatile solvents in such compositions. Moreover, the use of large amounts of volatile solvents in bitumen compositions may lower the viscosity of the resulting composition so that it no longer meets viscosity specifications designated for road paving  
30 applications.

U.S. Patent Nos. 3,803,066; 4,130,516; 4,145,322; 4,242,246; 5,019,610; and 5,371,121 are all hereby incorporated by reference for all purposes.

### SUMMARY OF THE INVENTION

The present invention provides a bitumen composition for road construction and repair, even when used without polymers. That bitumen composition comprises 75% to 99.95% of bitumen, 0 to 15% of a polymer, and 0.05% to 10% of an amide additive.

The amide additive is predominately an amide having the formula:



wherein  $R_1$  and  $R_2$  are alkyl groups, each having from 12 to 52 carbon atoms, and  $x$  is an integer of from 1 to 4.

- 10 This amide additive gives sufficient improvement so that polymer additives are not needed. By eliminating the use of polymers, one eliminates the need for sulfur treatment and the need for volatile solvents. Also, by not using polymers, it is easy to combine and blend the amide additive with base asphalt to make the asphalt binder. No complicated grinders or processing equipment are required. Since the blending is linear (i.e., the properties vary linearly with concentration, instead of exponentially), this allows for adjustment or optimization of properties easily and accurately.

- 20 Preferably, when polymers are not used, the bitumen composition comprises 95% to 99.75% of bitumen and 0.25% to 5% of amide additive. More preferably, it comprises 96% to 99.5% of bitumen and 0.5% to 4% of amide additive.

Preferably, when polymers are used, the bitumen composition comprises 85% to 99.4% of bitumen, 0.5% to 12% of polymer, and 0.1% to 3% of amide additive.

- 25 Preferably, the  $R_1$  and  $R_2$  alkyl groups are linear alkyl groups, each having from 12 to 20 carbon atoms, and  $x$  is 2. More preferably, the amide additive is predominately ethylene bis-stearamide.

A mixture can be formed having aggregate and from 2% to 8% of a bitumen composition of the present invention.

A storage stable, bitumen composition can be prepared by (a) combining 90% to 99.95% of bitumen and 0.05% to 10% of amide additive, and (b) mixing the product of step (a) at a temperature of from 250° F to 350° F for ½ to 1 hour.

- 5 A storage stable, bitumen composition can also be prepared by (a) combining 75% to 99.95% of bitumen, 0% to 15% of polymer, and 0.05% to 10% of amide additive, and (b) mixing the product of step (a) at a temperature of from 250° F to 400° F for ½ hour to 72 hours, depending upon the polymer and bitumen used.

- 10 Asphalt binders containing the proprietary mixture can have significantly lower viscosities at process and handling temperatures (135° C to 175° C) than polymer systems not containing the amide additive. This feature allows lower temperatures for plant storage, pumping, and field mixing. This characteristic could result in appreciable savings in energy requirements.

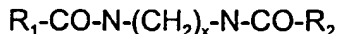
#### **BRIEF DESCRIPTION OF THE DRAWINGS**

- 15 In order to assist the understanding of this invention, reference will now be made to the appended drawings. The drawings are exemplary only, and should not be construed as limiting the invention.

- 20 Figure 1 shows a graph of the relationship between the predicted performance grade failure temperature of the asphalt as a function of amide content.

#### **DETAILED DESCRIPTION OF THE INVENTION**

- In its broadest aspect, the present invention involves a bitumen composition for road construction and repair. That composition comprises 75% to 99.95% of bitumen, 0 to 15% of polymer, and 0.05% to 10% of amide additive that is  
25 predominately an amide having the formula:



R<sub>1</sub> and R<sub>2</sub> are alkyl groups, each having from 12 to 52 carbon atoms, and x is an integer of from 1 to 4.

Prior to discussing the invention in further detail, the following terms will be defined:

### DEFINITIONS

As used herein the following terms have the following meanings unless  
5 expressly stated to the contrary:

The term "bitumen" (sometimes referred to as "asphalt") refers to all types of bitumens, including those that occur in nature and those obtained in petroleum processing, including air blown bitumen. The choice of bitumen will depend essentially on the particular application intended for the resulting  
10 bitumen composition.

The term "volatile solvent" refers to a hydrocarbon solvent that has a distillation end point less than or equal to 650° F. Such solvents are known to vaporize to some extent under ambient conditions and, accordingly, pose environmental concerns relating to hydrocarbon emissions.

15 The term "aggregate" refers to rock and similar material added to the bitumen composition to provide a mixture suitable for paving roads. Typically, the aggregate employed is rock indigenous to the area where the bitumen composition is produced. Suitable aggregate includes granite, basalt, limestone, and the like.

20 The "storage stable bitumen" refers to a bitumen composition that shows no evidence of skinning, settlement, gelation, or graininess and that the viscosity of the composition does not increase by a factor of four or more during storage at 325° ± 5° F (163° ± 2.8° C) for seven days. Preferably the viscosity does not increase by a factor of two (more preferably less than 50%) during  
25 that storage.

The term "predicted PG failure temperature," as used in the following examples, refers to the temperature at which a sample fails to meet the requirement of a minimum Dynamic Shear Rheometer (DSR) value of 1.0 kPa. Fundamental to PG binder testing is the fact that all PG grades have the  
30 same specification. It is only the temperature at which the binder is tested



which differentiates one grade from another. The high temperature PG grade requirement is defined as the temperature at which the Original DSR meets a minimum value of 1.0 kPa. By performing regression analysis on the data of Original DSR versus temperature one can then determine the temperature at which this particular sample would meet the minimum original value of 1.0 kPa, that temperature is the predicted PG failure temperature.

Unless otherwise specified, all percentages are in weight percent.

### THE POLYMER

There are three general types of polymers that are currently being used in the asphalt and road building industries: latex polymers, solid polymers, and ground-up automobile tire rubber. The most commonly used latex polymers are neoprene, SBR (styrene-butadiene-rubber), and natural rubber. The most commonly used solid polymers are SBR, EVA (ethylene-vinyl acetate), SB (styrene-butadiene), SBS (styrene-butadiene-styrene), and SIS (styrene-isoprene-styrene).

A preferred polymer is Elvaloy® AM, which is an epoxide-containing polymer produced by the process described in U.S. Patent No. 5,306,750, which is hereby incorporated by reference for all purposes.

### THE BITUMEN

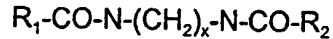
As stated above, the bitumen constitutes 75% to 99.95% of the bitumen composition.

Preferably, when polymers are not used, the bitumen constitutes from 95% to 99.75%, more preferably 96% to 99.5%, of the bitumen composition.

Preferably, when polymers are used, the bitumen constitutes from 85% to 99.4% of the bitumen composition.

## THE AMIDE ADDITIVE

The amide additive is an amide or mixture of amides and other additives, which is predominately an amide having the formula:



- 5      $R_1$  and  $R_2$  are alkyl groups, each having from 12 to 52 carbon atoms, and  $x$  is an integer of from 1 to 4. Preferably,  $R_1$  and  $R_2$  are linear alkyl groups, each having from 12 to 20 carbon atoms, and  $x$  is the integer 2. Most preferably, the amide additive is predominately ethylene bis-stearamide.

- 10     The preferred amide additive is the commercially available Kemamide® W-39, which is a fatty bisamide derived from stearic acid. Its major component is given in the publication "Opportunities with Humko Chemical Kemamide fatty amides" as  $RCONCH_2CH_2NCOR$ . That publication cites the use of a similar product (Kemamide® W-40) as a bisamide used in making asphalt/bisamide compounds useful in the production of potting and dipping compounds for  
15     electrical components.

Preferably, the amide additive constitutes from 0.05% to 10% of the bitumen composition, more preferably 0.25% to 5%, most preferably 0.5% to 4%.

## THE BITUMEN COMPOSITION

- 20     A storage stable, bitumen composition can be prepared by combining the bitumen and the amide additive, then mixing the composition at a temperature of from about 250° F to 350° F for about ½ to 1 hour.

- 25     A storage stable, bitumen composition can also be prepared by combining 75% to 99.95% of bitumen, 0% to 15% of polymer, and 0.05% to 10% of amide additive, then mixing that composition at a temperature of from 250° F to 400° F for ½ hour to 72 hours, depending upon the polymer and bitumen used. The bitumen composition is typically mixed with aggregate so as to provide a mixture suitable for use in paving roads. Typically, the aggregate composition comprises from 2% to 8% of bitumen composition with the balance of the composition being aggregate.

## EXAMPLES

The invention will be further illustrated by following examples, which set forth particularly advantageous method embodiments. While the Examples are provided to illustrate the present invention, they are not intended to limit it.

5

### EXAMPLES WITHOUT POLYMERS

#### COMPARATIVE EXAMPLE A NO AMIDE

A PBA-3 asphalt was determined to meet PG 58-28 requirements.

#### EXAMPLE 1

10 **1.50% AMIDE**

A 1500-gram batch of 98.50% of Comparative Example A and 1.50% Kemamide® W-39 was mixed at from 275° to 285° F for about 30 to 40 minutes, then tested at 64° C for initial Dynamic Shear Rheometer (DSR). Samples were packaged up into two 1-quart sample cans, sealed well, and  
15 stored overnight at 150° C. Full tests were made after overnight curing on one of the 1-quart samples. The other 1-quart sample was left in the oven (storage sample) for possible storage testing later. After overnight storage, the sample was tested for PG classification and was determined to meet PG 64-28 requirements.

20 **EXAMPLE 2**  
**2.00% AMIDE**

Another 1500-gram batch of Comparative Example A and Kemamide® W-39 was prepared according to the procedures of Example 1, except that the batch consisted of 98.00% of Comparative Example A and 2.00% Kemamide®  
25 W-39. After overnight storage, the sample was tested for PG classification and was determined to meet PG 70-28 requirements.

**EXAMPLE 3****2.50% AMIDE**

- Another 1500-gram batch of Comparative Example A and Kemamide® W-39 was prepared according to the procedures of Example 1, except that the batch consisted of 97.50% of Comparative Example A and 2.50% Kemamide® W-39. After overnight storage, the sample was tested for PG classification and was determined to meet PG 76-28 requirements.

**EXAMPLE 4****2.75% AMIDE**

- Another 1500-gram batch of Comparative Example A and Kemamide® W-39 was prepared according to the procedures of Example 1, except that the batch consisted of 97.25% of Comparative Example A and 2.75% Kemamide® W-39. After overnight storage, the sample was tested for PG classification and was determined to meet PG 76-22 requirements.
- The results of Comparative Example A and Examples 1 through 4 are summarized below:

**TABLE I**

Example	A	1	2	3	4
PBA-3	100.00%	98.50%	98.00%	97.50%	97.25%
Kemamide® W-39	0%	1.50%	2.00%	2.50%	2.75%
PG Grade	PG 58-28	PG 64-28	PG 70-28	PG 76-28	PG 76-22

- Figure 1 shows a graph of the relationship between the predicted performance grade failure temperature, as defined above, of the asphalt as a function of amide content. From Figure 1 and associated regression equation
- $$\text{Temperature} = 646.84(\% \text{ amide}) + 61.94$$
- it is easy to determine the quantity of Kemamide® W-39 that is required to meet the various PG Binder grades for that particular bitumen. Being linear makes it very easy to accurately blend these various grades either by blending up or blending down. For example, it requires about 1.25% Kemamide® W-39 blended into the asphalt of Comparative Example A to

meet minimum requirements for PG 70-XX. However, let's say we have a tank of PG 76-XX containing about 2.2% Kemamide® W-39 and want to blend it down to a PG 70-XX. We can do this by adding additional PBA-3, which would result in the final blend having about 1.25% Kemamide® W-39.

5

## EXAMPLES WITH POLYMERS

### COMPARATIVE EXAMPLE B NO POLYMER - NO AMIDE

A AC 20 asphalt was determined to meet PG 64-28 requirements.

### EXAMPLE 5

#### 10 NO POLYMER - 1.50% AMIDE

A 1500-gram batch was prepared according to the procedures of Example 1, except that the batch consisted of 98.50% of the AC 20 of Comparative Example B and 1.50% Kemamide® W-39. After overnight storage, the sample was tested for PG classification and was determined to meet PG 70-22 requirements (as compared to a performance grade of PG 64-28, without the amide).

15

### COMPARATIVE EXAMPLE C 2.25% POLYMER - NO AMIDE

A blend of 2.25% of Elvaloy® AM and 97.75% of the Comparative Example B was determined to meet PG 70-22 requirements.

20

### EXAMPLE 6

#### 2.25% POLYMER - 1.50% AMIDE

A 1500-gram batch was prepared according to the procedures of Example 1, except that the batch consisted of 2.25% of Elvaloy® AM, 96.25% of the Comparative Example B, and 1.50% Kemamide® W-39. After overnight storage, the sample was tested for PG classification and was determined to meet PG 76-22 requirements (as compared to a performance grade of PG 70-22, without the amide).

25

**COMPARATIVE EXAMPLE D**  
**2.50% POLYMER - NO AMIDE**

A blend of 2.50% of Elvaloy® AM and 97.50% of the Comparative Example B was determined to meet PG 70-22 requirements.

**5      EXAMPLE 7**

**2.50% POLYMER - 1.50% AMIDE**

10      A 1500-gram batch was prepared according to the procedures of Example 1, except that the batch consisted of 2.50% of Elvaloy® AM, 96.00% of the Comparative Example B, and 1.50% Kemamide® W-39. After overnight storage, the sample was tested for PG classification and was determined to meet PG 76-22 requirements (as compared to a performance grade of PG 70-22, without the amide).

15      Thus, in each instance, the use of the amide additive uniformly improves the performance grade of bitumens containing polymers. This data is summarized in the following table.

**TABLE II**  
**PERFORMANCE GRADE**

	No Amide	1.50% Amide
No Polymer	64-28	70-22
2.25% Polymer	70-22	76-22
2.50% Polymer	70-22	76-22

20      While the present invention has been described with reference to specific embodiments, this application is intended to cover those various changes and substitutions that may be made by those skilled in the art without departing from the spirit and scope of the appended claims.

What is claimed is:

1. A bitumen composition for road construction and repair, said composition comprising:
  - (a) 75% to 99.95% of a bitumen,
  - 5 (b) 0 to 15% of a polymer, and
  - (c) 0.05% to 10% of an amide additive, which is predominately an amide having the formula:
$$R_1-CO-N-(CH_2)_x-N-CO-R_2$$
wherein  $R_1$  and  $R_2$  are alkyl groups, each having from 12 to 52  
10 carbon atoms, and  $x$  is an integer of from 1 to 4.
2. A bitumen composition for road construction and repair according to Claim 1 wherein said composition is essentially free of polymer, and comprises from 95% to 99.75% of said bitumen and from 0.25% to 5% of said amide additive.
- 15 3. A bitumen composition for road construction and repair, according to Claim 2 wherein said bitumen composition comprises from 96% to 99.5% of said bitumen and from 0.5% to 4% of said amide additive.
4. A bitumen composition for road construction and repair according to Claim 1 wherein said composition comprises from 85% to 99.4% of said  
20 bitumen, from 0.5% to 12% of said polymer, and from 0.1% to 3% of said amide additive.
5. A bitumen composition for road construction and repair according to Claim 1 wherein said  $R_1$  and  $R_2$  alkyl groups are linear alkyl groups, each having from 12 to 20 carbon atoms, and  $x$  is 2.
- 25 6. A bitumen composition for road construction and repair according to Claim 5 wherein said amide additive is predominately an ethylene bis-stearamide.
7. An mixture comprising aggregate and from 2% to 8% of a bitumen composition for road construction and repair according to Claim 1.

8. A method for preparing a storage stable, bitumen composition which method comprises the steps of:

(a) combining:

- 5 (1) 90% to 99.95% of a bitumen, and  
(2) 0.05% to 10% of an amide additive, which is predominately an amide having the formula:



wherein  $R_1$  and  $R_2$  are alkyl groups, each having from 12 to 52 carbon atoms, and  $x$  is an integer of from 1 to 4, and

- 10 (b) mixing the product of step (a) at a temperature of from about 250° F to 350° F for about ½ to 1 hour.

9. A method for preparing a storage stable, bitumen composition which method comprises the steps of:

(a) combining:

- 15 (1) 75% to 99.95% of a bitumen, and  
(2) 0 to 15% of a polymer, and  
(3) 0.05% to 10% of an amide additive, which is predominately an amide having the formula:



20 wherein  $R_1$  and  $R_2$  are alkyl groups, each having from 12 to 52 carbon atoms, and  $x$  is an integer of from 1 to 4, and

- (b) mixing the product of step (a) at a temperature of from about 250° F to 400° F for about ½ to 72 hours.



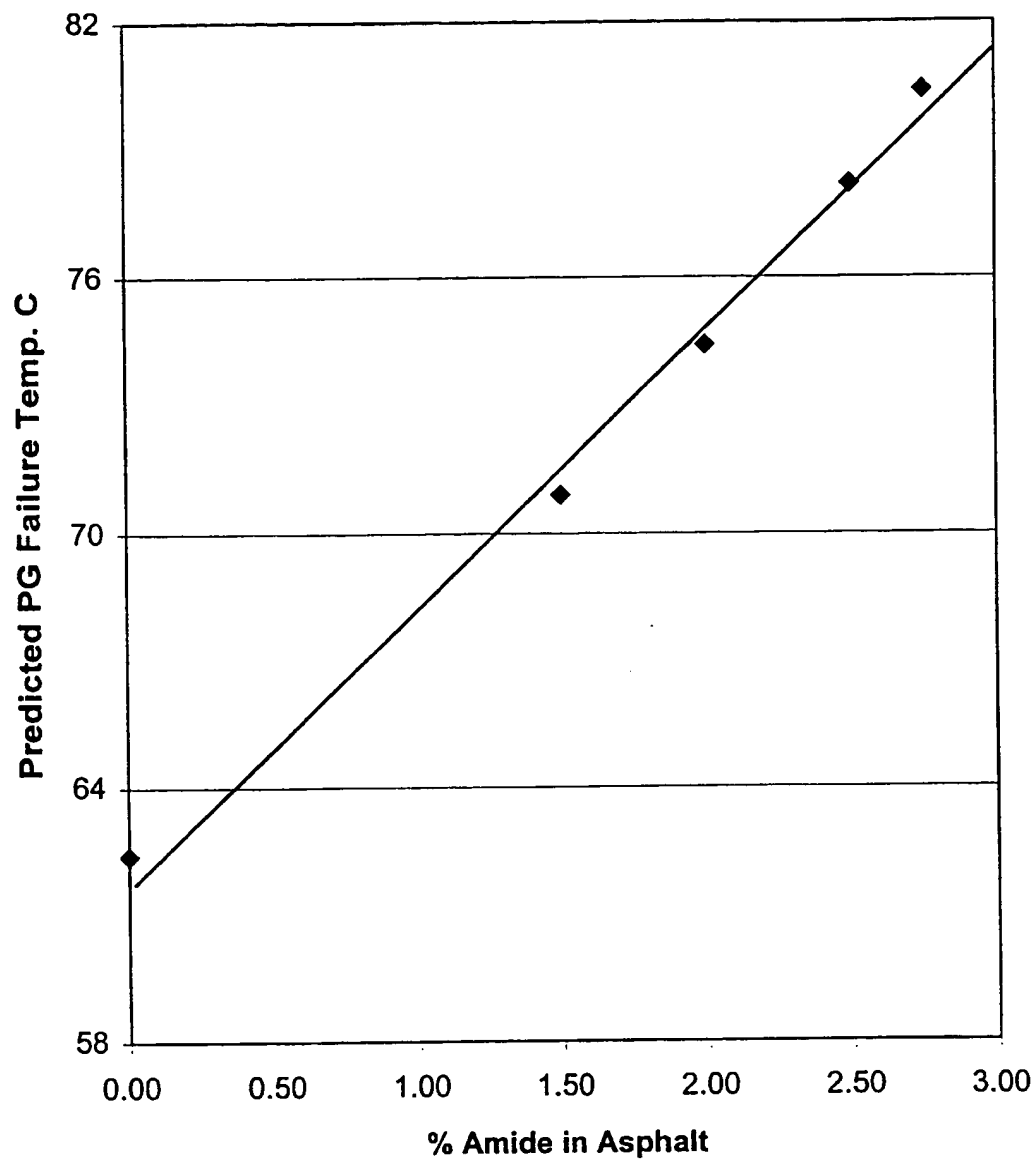


Figure 1

# INTERNATIONAL SEARCH REPORT

International Application No  
PCT/US 00/07287

**A. CLASSIFICATION OF SUBJECT MATTER**  
IPC 7 C08K5/20 C08L95/00

According to International Patent Classification (IPC) or to both national classification and IPC

**B. FIELDS SEARCHED**

Minimum documentation searched (classification system followed by classification symbols)  
IPC 7 C08K C08L

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data, PAJ, CHEM ABS Data

**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X Y	US 3 660 126 A (MORAN CHARLES E) 2 May 1972 (1972-05-02) column 2, line 10 - line 13 examples ---	1,2,5,6, 8,9 7
X Y	CA 1 260 653 A (OWENS CORNING FIBERGLASS CORP) 26 September 1989 (1989-09-26) claims 1,4 page 3, line 2 - line 15 ---	1-3,5,6 7
X	US 4 554 023 A (JANICKI RICHARD T) 19 November 1985 (1985-11-19) column 1, line 40 -column 2, line 10 ---	1-3,5,6
A	US 5 331 028 A (GOODRICH JOSEPH L) 19 July 1994 (1994-07-19) claims 1,17 --- -/--	1-9

☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

\* Special categories of cited documents :

- "A" document defining the general state of the art which is not considered to be of particular relevance
- "E" earlier document but published on or after the international filing date
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- "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
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Date of the actual completion of the international search

28 July 2000

Date of mailing of the international search report

11/08/2000

Name and mailing address of the ISA

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# INTERNATIONAL SEARCH REPORT

Inte. Jour. Classification No.

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